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Detection of anomalous Hall voltages in ultrahigh-mobility two-dimensional hole gases generated by optical spin orientation

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By combining optical spin orientation and an externally applied longitudinal electric field, transverse charge accumulation has been detected in very high-mobility two-dimensional hole gases by measuring the transverse voltage drop across simple Hall devices. Our results indicate intrinsic band-structure (rather than extrinsic skew scattering) derived spin-orbit coupling as the underlying mechanism of this spin-polarized transport effect.

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Recent years have seen a profusion of spin-polarized transport phenomena: the spin Hall effect (SHE) [1–4], anomalous Hall effect (AHE) [5], the inverse spin Hall effect (ISHE) [6], the spin-injection Hall effect (SIHE) [7], the spin galvanic effect [8], and the circular photogalvanic effect (CPGE) [9]. Such spintronic effects are of immense interest for quantum computing due to potential spintronic devices having faster switching times and lower power consumption than conventional electronic ones.

All of these above spintronic effects rely on the spin-orbit interaction (SOI) to bend the trajectories of spin-up and spin-down charge carriers in opposite directions. The spin-Hall conductivity thus arises as a result of the spin-orbit (SO) coupling, which can either be intrinsic to the band structure (Dresselhaus or Rashba), or alternatively, extrinsic in origin; deriving from asymmetric impurity scattering for up and down spins (skew scattering). There have been a number of reports of measurements, in both nonmagnetic semiconductors and metals, where the mechanism has been clearly extrinsic [2,4,6,7]. There have been very few claims, on the other hand, of measurements of a spin Hall conductivity, in nonmagnetic semiconductors, whose mechanism is intrinsic [3,10]. Although in Ref. [3] the authors state that theoretically the intrinsic mechanism *might* apply, they provide no experimental evidence and indeed their two-dimensional (2D) hole system is of very low mobility ($\mu = 3400\ \mathrm{cm^2/V\,s}$). Similarly in Ref. [10] the authors only speculate that *it is more likely* that their measurements of the photoinduced AHE of excitons in unstrained, undoped GaAs quantum wells at room temperature have an intrinsic origin, with no indication of the quality of their quantum wells given. Thus definitive detection of an *intrinsic* spin-polarized transport effect still remains elusive. Theoretical consensus is now that the intrinsic contribution is more likely to exist in p-type material and that it vanishes for n-type [11].

In inversion symmetric systems, the SHE, the AHE, and the ISHE (and thus the SIHE) are essentially the same phenomenon and there are established relations between them [12]. The drawback of the SHE [1–3] is that, due to the charge-balanced nature of its spin currents, electrical detection is impossible. In the AHE [5], on the other hand, the imbalance in the numbers of spin-up and spin-down carriers ensures that its transverse *spin* current generates a measurable transverse *charge* current or voltage. The AHE has traditionally been observed in ferromagnets and dilute magnetic semiconductors [5]. In nonferromagnetic materials, such as, for instance, nonmagnetic semiconductors, such a spin imbalance can either be generated by spin injection [4,6] or by optical spin orientation [7–10,13,14].

In this Rapid Communication we report the measurement of transverse spin voltages in ultrahigh-mobility 2D hole gases confined in nonmagnetic semiconductor heterostructures with low crystal inversion symmetry, where the spin-polarized current has been generated by the combination of optical spin orientation and a weak dc electric field.

There have been a couple of reports of the measurement of transverse spin voltages generated by optical spin orientation in nonmagnetic bulk semiconductors under extremely high longitudinal dc electric fields [13,14]—many orders of magnitude greater than those employed in the experiments reported in this Rapid Communication. In both, it appears that the AHE is extrinsic in origin. In Ref. [10] again high dc electric fields were employed. Although the authors speculate that their transverse spin voltage is intrinsic in origin, they have to invoke an "unavoidable built-in electric field and residual interface asymmetry" (for which they provide no evidence)

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to provide some Rashba SOI in their unstrained, undoped GaAs quantum wells. There has also been the report of the SIHE in a co-planar *pn* junction [7]—a similar device to that employed in Ref. [3]. In the SIHE work, a spin imbalance is produced by optical spin orientation and the results from the two-dimensional electron gas (2DEG) are explained as extrinsic in origin. The low-mobility ($\mu \simeq 3000\ \mathrm{cm^2/V\,s}$ [15]) 2D hole gas is located above the 2DEG, which is depleted in the dark, but when this side of the microdevice is illuminated, carriers can be photogenerated in both the 2D hole gas and its underlying 2DEG. Thus it is not clear whether the origin of the spin Hall conductivity, under such illumination, comes from the 2D hole gas or the 2DEG underneath.

In our ultrahigh-mobility–more than two orders of magnitude higher than in Refs. [3], [7] and [15]—symmetrically and asymmetrically silicon-doped (311)-grown GaAs-Al$_x$Ga$_{1-x}$As quantum wells (QWs), we have measured a transverse spin voltage (V_{TS}) that grows with the applied longitudinal electric field, the circular polarization of the incident light, and the 2D hole gas mobility—the latter we control with the temperature. Our results strongly indicate that the underlying mechanism is intrinsic in nature.

The (311)-grown GaAs-Al$_x$Ga$_{1-x}$As QWs were grown by molecular beam epitaxy (MBE). The symmetrically doped QW (symmetric QW) had a width of 30 nm and $x = 0.1$. For the asymmetrically doped well (asymmetric QW) these values were 15 nm and 0.33, respectively. Their 2D hole densities and mobilities, determined by Shubnikov–de Haas and quantum Hall measurements in the dark at 350 mK, were $p = 10^{11}\ \mathrm{cm^{-2}}$ and $\mu = 5 \times 10^5\ \mathrm{cm^2/V\ s}$ for the symmetric well and $p = 2.1 \times 10^{11}\ \mathrm{cm^{-2}}$ and $\mu = 9 \times 10^5\ \mathrm{cm^2/V\,s}$ for the asymmetric well. At 50 mK the mobility of the symmetric QW rises to $10^6\ \mathrm{cm^2/V\ s}$ [16], which corresponds to a momentum relaxation time of $\tau_p = 210$ ps ($\rightarrow \hbar/\tau_p = 3\ \mu$eV), assuming a hole effective mass $m^* = 0.37$. $p = 10^{11}\ \mathrm{cm^{-2}}$ corresponds to a Fermi energy of 0.65 meV $\equiv$ 7.5 K. Bulk GaAs has a SO splitting in its valence band of 0.34 eV. The symmetric QW sample was square with contacts in van der Pauw geometry, while the asymmetric QW sample was a Hall bar orientated along $[01\bar{1}]$.

We have also investigated both symmetrically and asymmetrically carbon-doped (100)-grown GaAs-Al$_x$Ga$_{1-x}$As QWs which were also grown by MBE ($x = 0.33$). Well widths were 15 and 20 nm, $p = 0.9 \times 10^{11}\ \mathrm{cm^{-2}}$ and $p = 2.0 \times 10^{11}\ \mathrm{cm^{-2}}$ and $\mu = 6.9 \times 10^5\ \mathrm{cm^2/V\ s}$ and $\mu = 4.3 \times 10^5\ \mathrm{cm^2/V\ s}$, respectively, measured in the dark at 350 mK. These (100)-grown samples were in the form of Hall bars.

All measurements were undertaken in a helium-3 cryomagnetic system with optical access via windows and magnetic fields up to 8 T, at temperatures ranging between 350 mK and 11 K. Circularly polarized optical excitation, perpendicular to the plane of the QWs, was achieved using a tunable continuous-wave Ti:sapphire laser, modulated by an electro-optical modulator (EOM) at 10 kHz, thereby producing a sinusoidal wave of left and right circularly polarized light. The Hall voltage was measured transversely to a dc current (3–6 μA) and in phase with the EOM. This ensures that any spin-unpolarized signals are eliminated [17]. The laser power density was varied up to 100 mW/cm^2. A schematic of the experimental arrangement is shown in Fig. 1(a), where σ indicates circularly polarized light.

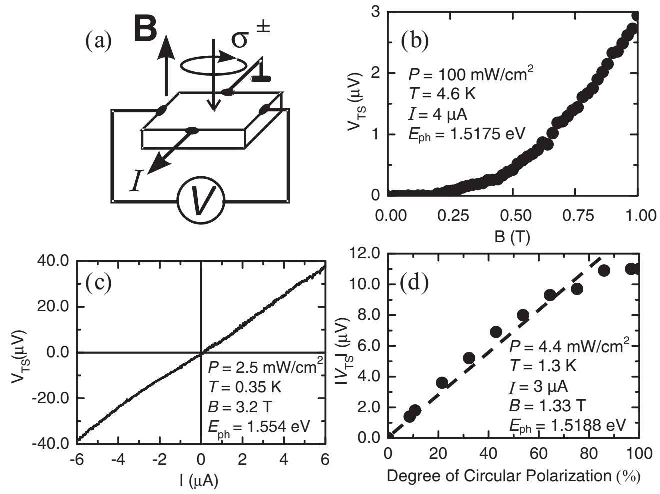

FIG. 1. (a) The experimental arrangement. (b) Magnetic field dependence of V_{TS} at low magnetic field for the symmetric QW. (c) The dc current dependence of V_{TS} for the asymmetric QW. (d) $|V_{\mathrm{TS}}|$ vs laser helicity for the symmetric QW. The dashed line is a guide for the eye.

Here we concentrate on the results from the (311)-grown QWs: Fig. 1(b) shows the magnetic field dependence of V_{TS} at very low magnetic field. It is clear that there is no measurable V_{TS} at zero field but that the signal grows dramatically with magnetic field. The perpendicular magnetic field spatially separates the photoexcited electron and hole *via* the classical Hall effect. If left unseparated, they would recombine or undergo spin relaxation (which is what one would expect for unseparated excitonic systems like those of Refs. [10] and [14]). Thus the magnetic field serves a similar purpose to the *pn* junction employed in Ref. [7], in that it spatially separates the electrons and holes. Illuminating a 2DEG or 2D hole gas in the absence of either a *pn* junction or a magnetic field does not produce a V_{TS} in these clean 2D systems [15]. In addition, a perpendicular magnetic field (parallel to the direction of light propagation) is known to increase V_{TS} further by suppression of Dyakonov-Perel spin relaxation [18]. This is discussed in more detail below.

We can distinguish V_{TS} from any conventional Hall voltage (V_{Hall}) because we are only measuring signals in phase with the EOM [17]. In fact typically, V_{TS} is two to three orders of magnitude smaller than V_{Hall} and thus attains maximum values of order 100 μV under our experimental conditions. This corresponds to transverse spin electric fields of order 10^{-2}–10^{-1} V/m and spin Hall angles $\alpha_H = \frac{V_{\mathrm{TS}}}{V_{xx}} \simeq 10^{-2}$–$10^{-1}$.

In Fig. 1(c) we show the linear dependence of V_{TS} on longitudinal current (I), as one would expect for such an anomalous Hall voltage. V_{TS} both passes through the origin and is symmetric about $I = 0$.

Figure 1(d) shows V_{TS} as a function of laser helicity. Zero polarization in Fig. 1(d) corresponds to the EOM switching between vertical and horizontal linearly polarized light, while 100% corresponds to the light flipping between left and right circular polarization. At all but the highest laser helicities, V_{TS} shows a linear dependence on the degree of circular

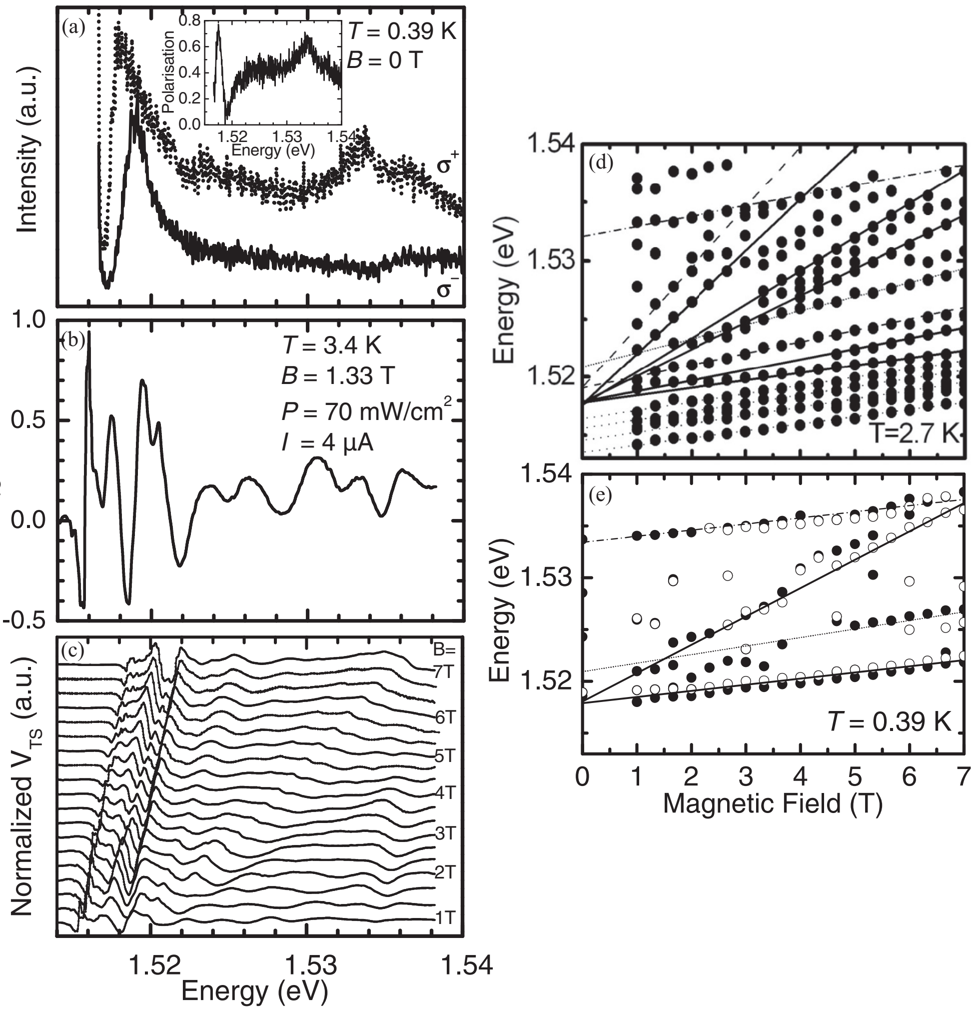

FIG. 2. (a) Zero magnetic field right (σ^+) and left (σ^-) circularly polarized PLE spectra for the symmetric QW. Inset: circular polarization of that PLE. (b) Photon energy dependence of V_{TS} for the same symmetric QW. (c) Photon energy dependence of that V_{TS} at various magnetic fields. (d) Magnetic field dependence of those V_{TS} peak energies. (e) Magnetic field dependence of σ^+ (solid circles) and σ^- (open circles) PLE peak energies for the same symmetric QW. The lines are guides for the eye and are explained in the text.

polarization of the laser, in analogy to the linear dependence on magnetization of the AHE in a ferromagnet. Unexpectedly, as the degree of circular polarization approaches 100% we observe some deviation away from a linear dependence and V_{TS} appears to saturate. This saturation behavior is not presently understood, however, it could be related to the suppression of the Dyakonov-Perel spin relaxation [18]: When a magnetic field is applied parallel to the optically aligned carrier spin orientation, the precession of the spins around the direction of the external magnetic field suppresses the precession around random internal magnetic fields and thereby inhibits spin relaxation. The rate of spin relaxation due to the Dyakonov-Perel mechanism starts to decrease when $\omega_c \tau_p \geqslant 1$ and saturates at $\omega_c \tau_p \gg 1$ and finally decreases to zero at $\omega_L \tau_p \gg 1$, where ω_c and ω_L are the cyclotron and Larmor frequencies, respectively [18]. At $B = 1$ T, $\omega_c \tau_p = 100$ and $\omega_L \tau_p = 133$, assuming an out-of-plane hole g factor of 7.2 [19] and $m^* = 0.37$. Thus $\omega_L \tau_p > \omega_c \tau_p \gg 1$. Only when the circular polarization of the laser gets close to 100% do we have sufficient carrier spin orientation for this effect to become noticeable. We have seen identical behavior in our CPGE measurements in magnetic field [20]. At zero magnetic field the dependence of the CPGE-induced V_{TS} on the laser helicity is strictly linear over the whole range from 0% to 100%. Thus this saturation behavior appears to be linked to the presence of the magnetic field.

We have taken circularly polarized photoluminescence excitation (circularly polarized PLE) spectra in both zero [Fig. 2(a)] and finite magnetic fields. With the detector set to an energy (1.516 eV) corresponding to the low-energy tail of the QW photoluminescence peak, the onset of PLE intensity coincides with that of the Fermi energy, which resides in the heavy-hole band. Thus the low-energy PLE peak, at 1.519 eV, in the zero magnetic field spectra of Fig. 2(a), is that of the ground-state light-hole to electron transition, which is most prominent in the left (σ^-) circularly polarized spectrum. The peak in Fig. 2(a) at 1.534 eV, which dominates in the PLE spectrum taken with right (σ^+) circularly polarized light, is the transition between the first-excited heavy hole and electron states. From these data, we have derived the polarization spectrum—inset of Fig. 2(a)—where the polarization is defined as $P = \frac{I_{\sigma^+} - I_{\sigma^-}}{I_{\sigma^+} + I_{\sigma^-}}$ and I_{σ^+} and I_{σ^-} are the photoluminescence intensities under σ^+ and σ^- excitation, respectively. In this polarization spectrum, the polarization minimum at 1.519 eV corresponds to the light-hole transition. We observe a maximum polarization of 75% at, what remains of, the ground-state heavy-hole to electron transition at 1.518 eV. We thus estimate that the hole Fermi sea is about 8% polarized [21]. However it should be noted that PLE measurements detect at the recombination energy, while our V_{TS} measurements involve polarized carriers at the Fermi energy.

We have measured V_{TS} as a function of photon energy in finite magnetic field. Figure 2(b) shows a typical spectrum. That V_{TS} has a value of zero at low energies indicates that these are below the absorption onset. Above this absorption onset the spectrum consists of a series of positive-voltage peaks and low- or negative-voltage troughs. We will show below that the sharp oscillations in V_{TS} at the very lowest energies can be assigned to transitions in the bulk GaAs. Those, generally broader, oscillations associated with the QW only occur above its absorption onset which is about 1.519 eV at $B = 1.33$ T. We have measured the photon-energy dependence of V_{TS} at various magnetic fields and this is shown in Fig. 2(c), where the spectra have been normalized for clarity as the V_{TS} signal grows rapidly with magnetic field.

At energies above 1.519 eV in Fig. 2(c) the V_{TS} oscillations spread out fanlike with increasing magnetic field in a manner reminiscent of the behavior of Landau levels. We have therefore plotted the V_{TS} peak energies versus magnetic field in Fig. 2(d). For comparison we have also plotted the circularly polarized PLE peak energies versus magnetic field in Fig. 2(e). It is clear that the fan chart derived from V_{TS} measurements in Fig. 2(d) is far richer than that derived from circularly polarized magneto-PLE spectra in Fig. 2(e), but comparison between the two allows clear identification of the transitions involved. In both Figs. 2(d) and 2(e) all lines are guides for the eye: the solid lines correspond to Landau levels associated with transitions between the heavy-hole and electron ground states (HH1-E1). Dashed lines correspond to ground-state light-hole transitions (LH1-E1). The dotted line indicates the forbidden HH3-E1 transition, while the dash-dotted line pinpoints the HH2-E2 transition. The widely spaced dotted lines below ~1.5175 eV in Fig. 2(d) do not appear in the QW PLE spectra of Fig. 2(e) and do not fan out and can thus be assigned to bulk GaAs excitons.

In Fig. 2(e) the PLE Landau levels do not appear to show significant spin splitting, whereas in Fig. 2(d)—the plot of V_{TS} peaks—there is clear Landau-level splitting—much greater than those expected for a bulk electron g factor of −0.44 [21]. In fact such splittings correspond, in this symmetric QW, to an out-of-plane heavy-hole g factor (g_z^{HH}) of 7 ± 2, which agrees remarkably well with the theoretically predicted value of g_z^{HH} of 7.2 [19]. However for our asymmetric QW similar V_{TS} peak Landau-level splittings correspond to $g_z^{\mathrm{HH}} = 15 \pm 3$. Thus the origin of these V_{TS} magnetically induced Landau-level splittings are not presently completely understood.

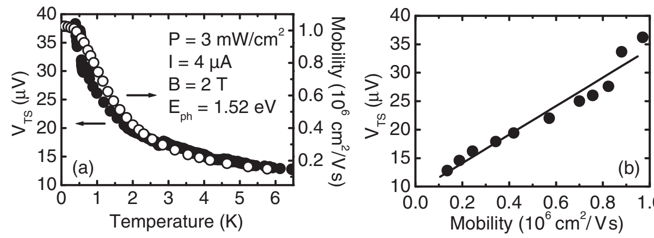

FIG. 3. (a) V_{TS} (filled circles) and 2D hole gas mobility [16] (open circles) for the same symmetric QW as a function of temperature. (b) V_{TS} vs mobility. The line is a guide for the eye.

V_{TS} is plotted versus temperature in Fig. 3(a). Also plotted in Fig. 3(a) is the temperature dependence of the 2D hole gas mobility of a sample taken from the same wafer [16]. It is obvious that the two curves in Fig. 3(a) show the same temperature dependence and so in Fig. 3(b) we have replotted the data as V_{TS} against mobility. There is a clear linear dependence. Due to the square shape of the symmetric QW sample, Fig. 3(b) does not pass through the origin as a result of unavoidable contributions to V_{TS} from the CPGE [9]. Similar considerations mean that the linear dependence of V_{TS} on I also does not pass through the origin for this square sample and explains why Fig. 1(c) shows data from the asymmetric QW, where the Hall bar is intentionally orientated along $[01\bar{1}]$ to eliminate just such CPGE contributions.

If the origin of V_{TS} were due to skew scattering, then according to theory, at absolute zero, $\alpha_H \propto 1/\sqrt{\mu}$ [7] and hence $V_{\mathrm{TS}} \propto \mu^{-3/2}$. Thus the linear dependence shown in Fig. 3(b) is a strong indication that the origin of V_{TS} in our case is not extrinsic. This conclusion is further reinforced as theory also states that the intrinsic contribution is not expected to dominate for $\beta k_{\mathrm{F}} \ll \hbar/\tau_p$ [7], where β is the Dresselhaus SO coupling coefficient strength—$\beta \approx -0.02$ eV Å [7]—and k_{F} is the Fermi wave vector. In our symmetric QW in fact $\beta k_{\mathrm{F}} \gg \hbar/\tau_p$ by at least a factor of 26. This thus puts us well into the intrinsic regime [22].

Comparison of the results from the asymmetric QW with those from the symmetric QW enables investigation of the effect of Rashba SO coupling [23]. We did not, however, detect any enhancement of V_{TS} in the asymmetric QW due to the additional Rashba contribution. For 2D electron gases in GaAs, the magnitude of the Rashba SO contribution is expected theoretically to be either equal to or up to ten times smaller than the Dresselhaus SO term [11], although the Rashba SO contribution for holes is expected to be greater than that for electrons.

Unexpectedly, our measurements of the very high-mobility C-doped (100)-grown 2D hole gases did not produce a photoinduced V_{TS} at any magnetic field. We can only surmise that this is due to the smaller Dresselhaus SO contribution in these high bulk inversion symmetry 2D hole gases, where $\beta \sim k^3$ (where k is the wave vector) compared to that in (311)-grown QWs, where $\beta \sim k$ ($k \ll 1$) [9].

In conclusion, our measurements of spin voltages, transverse to a weak dc electric field, generated by optical spin orientation in ultrahigh-mobility 2D hole gases in low crystal inversion symmetry semiconductor QWs, indicate that they are intrinsic rather than extrinsic in origin. We observe no enhancement of these intrinsic transverse spin voltages in 2D hole gases with additional Rashba SOI, induced by the asymmetry of their QWs. And, in contrast to our (311)-grown QWs, we were not able to measure any transverse spin voltages in very high-mobility 2D hole gases in high bulk inversion symmetry, (100)-grown C-doped QWs.

Note added. Our attention has been drawn to a recent publication reporting the valley Hall effect in monolayer MoS_2, where similar experimental techniques to those here are employed [24].

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[1] M. I. Dyakonov and V. I. Perel, Phys. Lett. A **35**, 459 (1971).
[2] Y. K. Kato, R. C. Myers, A. C. Gossard, and D. D. Awschalom, Science **306**, 1910 (2004).
[3] J. Wunderlich, B. Kaestner, J. Sinova, and T. Jungwirth, Phys. Rev. Lett. **94**, 047204 (2005).
[4] S. O. Valenzuela and M. Tinkham, Nature (London) **442**, 176 (2006).
[5] E. H. Hall, Philos. Mag. **10**, 301 (1880); **12**, 157 (1881); H. Ohno, H. Munekata, T. Penney, S. von Molnar, and L. L. Chang, Phys. Rev. Lett. **68**, 2664 (1992); T. Jungwirth, Q. Niu, and A. H. MacDonald, *ibid.* **88**, 207208 (2002).
[6] E. Saitoh, M. Ueda, H. Miyajima, and G. Tatara, Appl. Phys. Lett. **88**, 182509 (2006).
[7] J. Wunderlich, A. C. Irvine, J. Sinova, B. G. Park, L. P. Zârbo, X. L. Xu, B. Kaestner, V. Novák, and T. Jungwirth, Nat. Phys. **5**, 675 (2009).
[8] S. D. Ganichev, E. L. Ivchenko, V. V. Bel'kov, S. A. Tarasenko, M. Sollinger, D. Weiss, W. Wegscheider, and W. Prettl, Nature (London) **417**, 153 (2002).
[9] S. D. Ganichev, E. L. Ivchenko, S. N. Danilov, J. Eroms, W. Wegscheider, D. Weiss, and W. Prettl, Phys. Rev. Lett. **86**, 4358 (2001).
[10] J. L. Yu, Y. H. Chen, Y. Liu, C. Y. Jiang, H. Ma, L. P. Zhu, and X. D. Qin, Appl. Phys. Lett. **102**, 202408 (2013).
[11] J. Schliemann, Int. J. Mod. Phys. B **20**, 1015 (2006).
[12] P. Schwab, R. Raimondi, and C. Gorini, Europhys. Lett. **90**, 67004 (2010).
[13] M. I. Miah, J. Phys. D: Appl. Phys. **40**, 1659 (2007).
[14] N. Okamoto, H. Kurebayashi, K. Harii, Y. Kajiwara, H. Beere, I. Farrer, T. Trypiniotis, K. Ando, D. A. Ritchie, C. H. W. Barnes, and E. Saitoh, Appl. Phys. Lett. **98**, 242104 (2011).
[15] J. Wunderlich (private communication).
[16] A. P. Mills, A. P. Ramirez, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **83**, 2805 (1999).
[17] V. V. Bel'kov, S. D. Ganichev, P. Schneider, C. Back, M. Oestreich, J. Rudolph, D. Hägele, L. E. Golub, W. Wegscheider, and W. Prettl, Solid State Commun. **128**, 283 (2003).
[18] G. E. Pikus and A. N. Titkov, in *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya (North-Holland, New York, 1984).
[19] R. Winkler, S. J. Papadakis, E. P. DePoortere, and M. Shayegan, Phys. Rev. Lett. **85**, 4574 (2000).
[20] D. A. Vasyukov, A. S. Plaut, and M. Henini, Physica E **42**, 964 (2010).
[21] C. Weisbuch and C. Hermann, Phys. Rev. B **15**, 816 (1977).
[22] J. Sinova (private communication).
[23] E. I. Rashba, Physica E **34**, 31 (2006).
[24] K. F. Mak, K. L. McGill, J. Park, and P. L. McEuen, Science **344**, 1489 (2014).